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(54) Title: **IMPROVED BOTANICAL EXTRACTIONS PROCESS**

(57) Abstract: The present invention discloses a method of extracting biologically active compounds from botanic material, the method stabilising the botanical material from oxidative degradation. This preserves the biologically active compounds in the material. In the main embodiment, the plant material is mixed with a solution containing at least one acid and one antioxidant. It has been found that the oxidative degradation of the biologically active compounds in plant material can be prevented, slowed or stopped by the invention.

IMPROVED BOTANICAL EXTRACTION PROCESS

TECHNICAL FIELD

The present invention relates to an improved botanical extraction process. The invention
5 more specifically relates to an improved botanical extraction process that prevents degradation of biologically active compounds.

BACKGROUND ART

A number of biologically active compounds occur in flora, particularly the leafy portions,
10 in generally small quantities. For example, biologically active compounds (such as polyphenolics) in the leaf, roots, fruit and flowers can be extracted from *Echinacea*. Other compounds (alkamides, fructofuranosides and arabinogalactans) are also capable of extraction from *Echinacea*. Further, additional biologically active compounds (for
15 example bioflavonoids, polysaccharides, anti-oxidants, alkaloids, saponins, isoflavones, (etc)) are extracted commercially from many plant materials.

One key difficulty with commercial extraction of biologically active compounds is that the active compound(s) are unstable and, in particular, are prone to oxidation which
20 degrades the compounds in the extract.

One example of an extraction process is the solvent extraction of polyphenolics, fructofuranosides and alkamides from *Echinacea purpurea*. Most commercial extracts are prepared with an ethanol-water mixture which usually contains 25-70% ethanol.

25 Whilst solvent extraction techniques are more consistent in quality than water extraction techniques, they have the disadvantage that sometimes the solvents are undesired. Such solvents add to the processing cost, are sometimes difficult to handle, and can remove the 'natural' image of the product. Furthermore the use of solvents does not address the problem of oxidative degradation of the active compounds in the extract. Water
30 extraction techniques, whilst more 'natural' than use of solvents, can be less successful at extraction.

Degradation of the biologically active compounds occurs via a number of possible oxidation reactions. It is generally thought that polyphenolics and the polysaccharides found in, for example, *Echinacea* plant material provide part of the medicinal activity. As these polyphenolics are released in the extraction process, enzymes and polyphenolic oxidase (PPO) in particular, cause an oxidation reaction to occur. The PPO enzyme appears to be highly active in *Echinacea* with research showing an almost 80% reduction in phenolic levels within 5 minutes. Other oxidation reactions also occur but the above is the fastest to degrade the biological compounds of the extract

10 This degradation of biologically active compounds during extraction is sometimes further exacerbated by degradation during drying and/or storage of plant material prior to the extraction process, when fresh material is not used in the extraction process.

US6,217,878 attempts to address the problem of oxidative degradation by utilising heat at the start of the process to denature the PPO enzymes causing the oxidation reaction. This is a blanching type operation. While heat destroys the enzyme which catalyses the oxidation reaction, further problems are caused including heat degradation of the phenolic compounds and other active compounds. As a result, final phenolic activity is reduced. The invention also requires the use of a carrier compound (tricalcium phosphate) in reasonably large quantities thus introducing new compounds to the method.

The above discussion has been directed to that of *Echinacea* extraction. It will be appreciated by those skilled in the art that the principles of degradation of biologically active compounds in all plant material are well illustrated by this plant.

25 It is an object of the present invention to address the above problems by providing a method of extraction of compounds from plant material that avoids oxidative degradation of biologically active compounds in the plant material.

30 It is a further object of the present invention to provide a method of extraction that is cost effective.

It is still a further object of the invention to disclose a method that addresses pH control in extract processing.

- 5 It is an object of the present invention to address the foregoing problems or at least to provide the public with a useful choice.

All references, including any patents or patent applications cited in this specification are hereby incorporated by reference. No admission is made that any reference constitutes
10 prior art. The discussion of the references states what their authors assert, and the applicants reserve the right to challenge the accuracy and pertinency of the cited documents. It will be clearly understood that, although a number of prior art publications are referred to herein, this reference does not constitute an admission that any of these documents form part of the common general knowledge in the art, in New Zealand or in
15 any other country.

It is acknowledged that the term 'comprise' may, under varying jurisdictions, be attributed with either an exclusive or an inclusive meaning. For the purpose of this specification, and unless otherwise noted, the term 'comprise' shall have an inclusive meaning - i.e. that it will be taken to mean an inclusion of not only the listed components
20 it directly references, but also other non-specified components or elements. This rationale will also be used when the term 'comprised' or 'comprising' is used in relation to one or more steps in a method or process.

Further aspects and advantages of the present invention will become apparent from the
25 ensuing description which is given by way of example only.

Ascorbic acid is a white, crystalline vitamin, $C_6H_8O_6$, found in citrus fruits, tomatoes, potatoes, and leafy green vegetables and used to prevent scurvy. It is also called vitamin C. It is known as an antioxidant in the food industry.

Citric acid is an organic acid containing three carboxyl groups and has the chemical formula of $C_6H_8O_7$. It is found in citrus fruits (oranges, grapefruit, lemons). It is used in commercially processed foods and can complex some metals that act as catalysts in oxidation.

5

DISCLOSURE OF INVENTION

According to one aspect of the present invention there is provided a method of extracting biologically active compounds from botanical material, said method including the step of:

- 10 (a) mixing together the botanical material and a solution including at least one acid and at least one antioxidant to form a mixture;
- characterised in that the combination of an acid and an antioxidant substantially prevents, slows and/or halts oxidative degradation of compounds in the mixture.

- 15 According to a further aspect of the present invention there is provided a method of extracting biologically active compounds from botanical material substantially as described above, said method further characterised in that the use of the solution in step (a) prevents, slows and/or halts polyphenolic oxidation reactions in the mixture.

- 20 According to a further aspect of the present invention there is provided a method of extracting biologically active compounds from botanical material substantially as described above, said method further characterised in that the use of the solution in step (a) reverses the polyphenolic oxidation reaction equilibrium.

- 25 According to another aspect of the present invention there is provided a method of extracting biologically active compounds from botanical material substantially as disclosed above, wherein the solution of step (a) is added to the botanical material by means selected from the group: spraying; dipping; pouring over; and a combination thereof.

- 30 According to a further aspect of the present invention there is provided a method of extracting biologically active compounds from botanical material substantially as

described above, wherein the solution in step (a) is food grade quality. Preferably, the solution is a mixture selected from the group including: citric acid; ascorbic acid; cysteine; cinnamic acid; sulphur dioxide; vitamin E; and any combination thereof.

- 5 According to a further aspect of the present invention there is provided a method of extracting biologically active compounds from botanical material substantially as described above, wherein the solution in step (a) includes hexyl resorcinol

- 10 According to a further aspect of the present invention there is provided a method of extracting biologically active compounds from botanical material substantially as described above, wherein the solution of step (a) is: 0.5-30% (by weight) citric acid; 0.5-30% (by weight) ascorbic acid; and water. Optionally, the acid components are added as a powder or a prepared liquid.

- 15 According to a further aspect of the present invention there is provided a method of extracting biologically active compounds from botanical material substantially as described above, wherein the solution in step (a) includes water that has been deoxygenated and/or chilled to between 1 and 4°C.

- 20 According to a further aspect of the present invention there is provided a method of extracting biologically active compounds from botanical material substantially as described above, wherein the material is selected from the range of plants known to be used for herbal medicines and natural remedies. Preferably, said plant material is prone to oxidative reactions that reduce the activity of the material.

25

Most preferably, said botanical material is selected from the group including: *Echinacea*; Ginkgo Biloba; Kava Kava; Ginseng; Black Cohosh; Green Tea; St John's Wort (with hypercerin as an active); Artichoke; Chamomile; Dong Quai; Grape Seed; Grape Skins; berries; Hawthorn; Hops; Passion Flower; Pine Bark; Red Clover; Olive Leaf; currants;

- 30 and combinations thereof.

Preferably, the material selected is the leafy portion of a plant, free of stems and branches and/or twigs. It be appreciated, however, that any part of the plant, including flower heads, or roots may be used.

- 5 Optionally, the botanical material is the by product or pressate of other plant processing/extraction steps. Optionally, the plant material is the by product of, for example, a seed extraction, a super critical fluid extraction (SCFE), or the by product of flower harvesting.
- 10 According to a further aspect of the present invention there is provided a method of extracting biologically active compounds from botanical material substantially as described above, wherein step (a) occurs immediately before harvest of the plant material.

- According to a further aspect of the present invention there is provided a method of
15 extracting biologically active compounds from botanical material substantially as described above, said method including a step before step (a) of:

(xa) disintegrating the botanical material.

- Preferably, the step of disintegration occurs within a time of less than 5 minutes before
20 step (a). Most preferably this time is less than 10 seconds.

- According to a further aspect of the present invention there is provided a method of
extracting biologically active compounds from botanical material substantially as
described above, wherein step (xa) is simultaneous with step (a).

25

In a preferred embodiment, the plant material disintegrated is fresh material. Optionally, the material is frozen prior to disintegration.

- Preferably, the botanical material is disintegrated to a size of 0.1mm to 50mm in length.
30 Preferably, disintegration is completed by a method selected from the group including: rubbing; milling; chopping; and combinations thereof.

According to a further aspect of the present invention there is provided a method of extracting biologically active compounds from botanical material, said method further including the step, after step (a) of:

- 5 (b) separating the liquid from the residue;

Preferably, step (b) is completed at any time between immediately after step (a) and 7 days after step (a).

- 10 Optionally also, during the contact time period described above, the botanical mixture is periodically or continuously stirred, mixed or otherwise agitated.

- 15 According to another aspect of the present invention there is provided a method of extracting biologically active compounds from botanical material substantially as disclosed above, wherein step (b) is completed by: collecting a liquid extract as the retentate of reverse osmosis; collecting a liquid extract as the retentate of ultra-filtration, or collecting a liquid extract as the retentate of a combination of these methods.

- 20 According to another aspect of the present invention there is provided a method of extracting biologically active compounds from botanical material substantially as disclosed above, wherein in step (b) the method of separation to collect a liquid extract is selected from the group including: filtration; super critical fluid extraction (SCFE); mechanical de-watering; and any combination thereof.

- 25 Most preferably the method of separation is a combination of filtration and mechanical de-watering.

- 30 According to a further aspect of the present invention there is provided a method of extracting biologically active compounds from botanical material substantially as described above wherein said method includes a further step, after step (b) of:

(c) drying the liquid extract from step (b) to obtain a solid extract.

According to another aspect of the present invention there is provided a method of extracting biologically active compounds from botanical material substantially as described above, wherein said method includes a further step, after step (b) of:

- 5 (d) pressing the residual plant material to obtain a pressate which is then combined with the liquid extract of step (b).

According to another aspect of the present invention there is provided a method of extracting biologically active compounds from botanical material substantially as described above, wherein said method includes a further step, after step (d) of:

- 10 (e) filtrate and pressate drying.

The filtrate and or pressate (if present) are preferably dried by freeze drying, spray drying or other known drying techniques.

15 According to a yet further aspect of the present invention there is provided a method of extracting biologically active compounds from botanical material substantially as described above, wherein any one or a combination of steps described above are conducted in the absence of oxygen. Optionally, a nitrogen atmosphere is used.

20 According to a yet further aspect of the present invention there is provided a method of extracting biologically active compounds from botanical material, as claimed in any preceding claim, wherein the above described method includes the further step, after step (a) and the sequences of steps selected from steps (b), (c), (d), or (e), said step being:

- 25 (f) one or more further cycles of at least one of steps (a), (b), (c), (d), or (e).

According to a yet further aspect of the present invention there is provided a method of extracting biologically active compounds from botanical material substantially as described above, wherein the steps up to and including obtaining the liquid extract are conducted at the harvest site.

30

According to a further aspect of the present invention there is provided a method of extracting biologically active compounds from botanical material substantially as described above, wherein the phenolic level after extraction is maintained at 50–100% as compared with the starting levels, measured as dried extract. Preferably, the phenolic level after extraction is maintained at 70–100% of starting levels.

According to another aspect of the present invention there is provided a product formed by the process as described above wherein the product is the mixture from step (a).

10 According to another aspect of the present invention there is provided a product formed by the process as described above wherein the product is the liquid extract from step (b).

According to another aspect of the present invention there is provided a product formed by the process as described above wherein the product is the residue from step (b).

15

According to another aspect of the present invention there is provided a product formed by the process as described above wherein the product is the mixture from step (d).

20 According to another aspect of the present invention there is provided a product formed by the process substantially as described above, wherein the product of the process is a solid.

It has been found to be advantageous to use only the leafy part of botanical material with the above method for extraction of polyphenolics. However, it is to be noted that 25 biologically active compounds can concentrate in any part of a plant. Thus the selection of a part of a plant to be extracted advantageously over others, will be dependent upon the compound to be extracted(s) and the plant from which it is extracted.

It will be known to those skilled in the art that biologically active compounds can 30 deteriorate if biological material is dried or is picked in poor condition etc. The use of the above described method, by taking botanical material direct from harvest as described,

ensures that there is very little opportunity for degradation of the biologically active compounds.

The advantage of the invention as described above is that the oxidative degradation of the biologically active compounds in plant material can be prevented or stopped or slowed as soon as the degradation starts to occur.

Aspects of the present invention will be apparent from the following examples which are given by example only.

10

BEST MODES FOR CARRYING OUT THE INVENTION

Example 1

Take 400kg of frozen *E. purpurea* tops and pass through a granulator and re-freeze. Note that freezing is not essential.

15

Prepare solution by mixing 160g of citric acid with 220g of ascorbic acid, dilute with water to 1000ml and mix thoroughly.

Example 1.1

Take 10kg of frozen *E. purpurea* and spray with 1L of solution. Mix together and thaw to 15°C or more. Add 19 litres of water to the mixture and stand with occasional stirring for approximately 2 hours.

20

Using a filter bag, filter the mixture and separate out the solid and liquid extract. Optionally, freeze the liquid extract for storage at this point or continue into the next step of evaporation to concentrate up to 50% total solids. Freeze down the remaining extract and freeze dry to collect a final powdered *Echinacea* extract.

25

Example 1.2

Take 5kg of frozen granulated *E. purpurea* tops and mince. Add 500ml of solution prepared as above.

30

Add water, press and filter as in Example 1.1 above. Freeze the collected extract and freeze dry.

5 **Example 1.3**

Take 1kg of frozen granulated *E. purpurea* tops and blend. Add 100ml of solution and mix thoroughly.

10 Add water, press and filter as in Example 1.1 above. Freeze the collected extract and freeze dry.

Example 2

25 tonnes of raw *E. purpurea* material is added into a mill. The mill of known construction, mills or chops the raw *Echinacea* bringing particles down to 10-20 mm,
15 followed by a rubbing action mill similar to that used for grain.

As soon as particles emerge from the mill they are sprayed with a citric and ascorbic acid solution. The solution is a mixture of 400kg of citric acid, 550kg of ascorbic acid and then diluted using water. A small amount of thickener may optionally be added to the
20 solution to ensure that the solution 'sticks' to the *Echinacea* material.

After this, the *Echinacea*/acid mixture can be further processed safely via known techniques including as pressing, filtration, pasteurisation, chilling, freezing, evaporation and drying.
25

In the above process a 2.5% starting phenolic level (dry basis by weight by HPLC analysis) is concentrated up to a 75% phenolic recovery. A 6.8% fructofuranoside level (dry basis by weight) is concentrated up to an 80% recovery as well

30 **Example 3**

Comparison of phenolic activity from standard water extraction methods without acid

treatment compared to acid treatment methods.

The water extraction method was used as a control measure. Water extraction was used as per the methods above but without the addition of any citric or ascorbic acids.

5

Citric and ascorbic acids were added to the dilution water used to submerge the plant material before disintegration. The concentration of citric acid in the dilution water was 0.64 wt %, while that for ascorbic acid was 0.18 or 0.88 wt %, respectively. The plant slurry contained 6.3% dry plant material. The final pH of the extract ranged from 3.6 to

10 4.

The results were as follows:

Sample	Extract yield (% of dry plant)	Phenolics in extract (% w/w of extract)			Yield of phenolics (% of phenolics in plant compared with base levels)		
		Caftaric	Cichoric	Total	Caftaric	Cichoric	Total
Water (control)	35	0.03	0.06	0.09	2	1	1
Citric acid (33.3mM)	46	0.9	1.0	1.9	83	28	41
Citric (33.3mM) + Ascorbic acid (50mM)	60	1.6	3.4	4.9	179	122	134

15 Notes:

- Results based on HPLC analysis
- Freeze-dried extracts were analysed
- Freeze-dried plant contained 0.5% caftaric acid and 1.7% cichoric acid, thus a total of 2.2% phenolics

20

As is demonstrated above the phenolic levels in the extract are higher for citric acid alone and significantly higher for the citric and ascorbic acid combination. Similarly the

percentage yield of phenolics in the plant are significantly higher, particularly for the citric and ascorbic acid combination where levels above 100% recovery are achieved. This suggests that the oxidation reaction is being reversed and oxidative phenolics in the extract initially are being reversed back to un-oxidised phenolics. It is appreciated that the combination of citric and ascorbic acids creates a synergistic mixture that greatly enhances the phenolic yield in botanical extracts. The mixture also co-extracts fructofuranosides effectively.

Aspects of the present invention have been described by way of example only and it should be appreciated that modifications and additions may be made thereto without departing from the scope thereof as defined in the appended claims.

WHAT WE CLAIM IS:

1. A method of extracting biologically active compounds from botanical material, said method including the step of:
 - (a) mixing together a botanical material and a solution including at least one acid and at least one antioxidant to form a mixture;characterised in that the combination of an acid and an antioxidant substantially prevents, slows and/or halts oxidative degradation of compounds in the mixture.
2. A method of extracting biologically active compounds from botanical material as claimed in claim 1, said method further characterised in that the use of the solution in step (a) prevents, slows and/or halts polyphenolic oxidation reactions in the mixture.
3. A method of extracting biologically active compounds from botanical material as claimed in claim 1 or claim 2, said method further characterised in that the use of the solution in step (a) reverses the polyphenolic oxidation reaction equilibrium.
4. A method of extracting biologically active compounds from botanical material as claimed in any one of the above claims, wherein the solution of step (a) is added to the botanical material by means selected from the group: spraying; dipping; pouring over; and a combination thereof.
5. A method of extracting biologically active compounds from botanical material as claimed in any one of the above claims, wherein the solution in step (a) is food grade quality.
6. A method of extracting biologically active compounds from botanical material as claimed in any one of the above claims, wherein the solution is a mixture selected from the group including: citric acid; ascorbic acid; cysteine; cinnamic acid; sulphur dioxide; vitamin E; and any combination thereof.

7. A method of extracting biologically active compounds from botanical material as claimed in any one of the above claims, wherein the solution in step (a) includes hexyl resorcinol
8. A method of extracting biologically active compounds from botanical material as claimed in any one of the above claims, wherein the solution of step (a) is: 0.5-30% (by weight) citric acid; 0.5-30% (by weight) ascorbic acid; and water.
9. A method of extracting biologically active compounds from botanical material as claimed in any one of the above claims, wherein the solution in step (a) includes water that has been deoxygenated and/or chilled to between 1 and 4°C.
10. A method of extracting biologically active compounds from botanical material as claimed in any one of the above claims, wherein said plant material is prone to oxidative reactions that reduce the activity of the biologically active material.
11. A method of extracting biologically active compounds from botanical material as claimed in any one of the above claims, wherein said botanical material is selected from the group including: *Echinacea*; Ginkgo Biloba; Kava Kava; Ginseng; Black Cohosh; Green Tea; St John's Wort (with hypercerin as an active); Artichoke; Chamomile; Dong Quai; Grape Seed; Grape Skins; Hawthorn; Hops; Passion Flower; Pine Bark; Red Clover; Olive Leaf; currants; berries; and a combination thereof.
12. A method of extracting biologically active compounds from botanical material as claimed in any one of the above claims, wherein the material selected is the leafy portion of a plant, free of stems and branches and/or twigs.
13. A method of extracting biologically active compounds from botanical material as claimed in any one of the above claims, wherein the botanical material is the by product or pressate of other plant processing/extraction steps.

14. A method of extracting biologically active compounds from botanical material as claimed in claim 13, wherein the plant material is the by product of: a seed extraction; a super critical fluid extraction (SCFE); the by product of flower harvesting.
15. A method of extracting biologically active compounds from botanical material as claimed in any one of claims 4 to 14, wherein step (a) occurs immediately before harvest of the plant material.
16. A method of extracting biologically active compounds from botanical material as claimed in any one of the above claims, said method includes a step of:
(xa) disintegrating the botanical material.
17. A method of extracting biologically active compounds from botanical material as claimed in claim 16, wherein the step of disintegration occurs within a time of less than 5 minutes of step (a).
18. A method of extracting biologically active compounds from botanical material as claimed in claim 16 or claim 17, wherein the step of disintegration occurs within a time of less than 10 seconds of step (a).
19. A method of extracting biologically active compounds from botanical material as claimed in any one of claims 16 to 18, wherein the step of disintegrating the botanical material is simultaneous with step (a).
20. A method of extracting biologically active compounds from botanical material as claimed in any one of claims 16 to 19, wherein the botanical material is disintegrated fresh material.
21. A method of extracting biologically active compounds from botanical material as claimed in any one of claims 16 to 20, wherein the botanical material is disintegrated frozen material.

22. A method of extracting biologically active compounds from botanical material as claimed in any one of claims 16 to 21, wherein the botanical material is disintegrated to a size of 0.1mm to 50mm in length.

23. A method of extracting biologically active compounds from botanical material as claimed in any one of the above claims, wherein said method further includes the step, after step (a) of:

(b) separating the liquid from the residue;

24. A method of extracting biologically active compounds from botanical material as claimed in claim 23, wherein step (b) is completed at any time between immediately after step (a) and 7 days after step (a).

25. A method of extracting biologically active compounds from botanical material as claimed in either claim 23 or 24, wherein in step (b) the method of separation is selected from the group including: filtration; super critical fluid extraction (SCFE); mechanical de-watering; and a combination thereof.

26. A method of extracting biologically active compounds from botanical material as claimed in any one of claims 23 to 25, wherein the extraction process in step (b) is filtration and mechanical de-watering in combination.

27. A method of extracting biologically active compounds from botanical material as claimed in either claim 23 or 24, wherein step (b) is completed by: collecting a liquid extract as the retentate of reverse osmosis; collecting a liquid extract as the retentate of ultra-filtration, or collecting a liquid extract as the retentate of a combination of these methods.

28. A method of extracting biologically active compounds from botanical material as claimed in any one of claims 23 to 27, wherein said method includes a further step after

step (b) of:

(c) drying the liquid extract from step (b) to obtain a solid extract.

29. A method of extracting biologically active compounds from botanical material as claimed in any one of claims 23 to 27, wherein said method includes a further step after step (b) of:

(d) pressing the residual plant material to obtain a pressate which is then combined with the liquid extract of step (b).

30. A method of extracting biologically active compounds from botanical material as claimed in claim 29, wherein said method includes a further step after step (d) of:

(e) filtrate and pressate drying.

31. A method of extracting biologically active compounds from botanical material as claimed in any one of the above claims, wherein any one or a combination of steps described above are conducted in the absence of oxygen.

32. A method of extracting biologically active compounds from botanical material as claimed in any one of the above claims, wherein a nitrogen atmosphere is used.

33. A method of extracting biologically active compounds from botanical material, as claimed in any preceding claim, wherein the above described method includes the further step, after step (a) and the sequences of steps selected from steps (b), (c), (d), or (e), said step being:

(f) one or more further cycles of at least one of steps (a), (b), (c), (d), or (e).

34. A method of extracting biologically active compounds from botanical material as claimed in any one of the above claims, wherein the phenolic level after extraction is maintained at 50–100% as compared with the starting levels, measured as dried extract.

35. A method of extracting biologically active compounds from botanical material as

claimed in claim 34, wherein the phenolic level after extraction is maintained at 70–100% of starting levels.

36. A product formed by the process as claimed in any preceding claim wherein the product is the mixture from step (a).
37. A product formed by the process as claimed in any one of claims 23 to 35 wherein the product is the extract from step (b).
38. A product formed by the process as claimed in any one of claims 23 to 35 wherein the product is the residue from step (b).
39. A product formed by the process as claimed in any one of claims 29 to 35 wherein the product is the mixture from step (d).
40. A product formed by the process as claimed in any preceding claim, wherein the product of the process is a solid.
41. A method of extracting biologically active compounds from botanical material substantially as claimed in any one of claims 1 to 35, as hereinbefore described and with reference to the accompanying examples.
42. A product formed by the process substantially as claimed in claim 36, as hereinbefore described and with reference to the accompanying examples.
43. A product formed by the process substantially as claimed in claim 37, as hereinbefore described and with reference to the accompanying examples.
44. A product formed by the process substantially as claimed in claim 38, as hereinbefore described and with reference to the accompanying examples.

45. A product formed by the process substantially as claimed in claim 39, as hereinbefore described and with reference to the accompanying examples.

46. A product formed by the process substantially as claimed in claim 40, as hereinbefore described and with reference to the accompanying examples.

INTERNATIONAL SEARCH REPORT

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A. CLASSIFICATION OF SUBJECT MATTERInt. Cl. ⁷: A23F 3/16, 3/18; C07B 63/00; C07C 39/19, 39/21, 57/42, 59/52; C07G 17/00

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B. FIELDS SEARCHED

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Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

STN FILE CA, WPIDS, JAPIO, MEDLINE Keywords: echinacea, ginkgo, kava, ginseng cohosh, green tea, wort, artichoke, chamomile, grape, berry, pine, citric, ascorbic, cysteine, cinnamic, sulfur(W) dioxide, sulphur(W)dioxide, vitamin E, tocopherol, extract, phenol

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 6004559 A (SINNOT, R A et al) 21 December 1999 See claim 1	1-5, 13
X	Derwent Abstract Accession No. 96-441503/44, Class D16, RU 2053270-C1 (OENOLAB CO LTD) 27 January 1996 See example	1-5, 10, 13
X	US 5427806 A (EKANAYAKE, A et al) 27 June 1995 See abstract, column 1 line 24 to column 2 line 62; column 4 lines 1 to column 6 line 14; column 7 line 35-47; claims 1, 8 and 14	1-6, 8, 10-12, 16-20, 22-24, 29-33, 36-40

☒ Further documents are listed in the continuation of Box C
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"O" document referring to an oral disclosure, use, exhibition or other means	
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Date of the actual completion of the international search
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INTERNATIONAL SEARCH REPORT

International application No.

PCT/NZ02/00230

C (Continuation).

DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	AUBERT, S et al "Anthocyanins stability of apple purees", Bulletin de Liason - Groupe Polyphenols 1992 vol 16 part 2 pages 52-55. See "Resume" lines 6-8; "Summary"; Figure 3; "Conclusions"	1-6, 10, 13, 36, 40
X	CANTARELLI, C et al "Stabilization of pome and grape juice against phenolic deterioration by enzymatic treatments", Internationale Fruchtsaft-Union, Wissenschaftlich-technische Kommission, [Berichte] 1990 vol 21 pages 35-57. See page 35 paragraphs 4-5, page 37 paragraph 1, page 47 paragraph 4, Figure 6	1-6, 10, 13, 34
X	STN FILE CA, Abstract No. 104:67831 & JP 60192548 (SUNTORY, LTD) 1 October 1985 See abstract	1-46
X	Derwent Abstract Accession No. 83-03555k, Class D13 E19, JP 57 194749 (KATO K) 30 November 1982 See abstract	1-46
X	STN FILE CA, Abstract No. 78:2710 & IVANYUTINA, A I, Sadovodstvo, Vinogradarstvo i Vinodelie Moldavii (1954-1986) (1972) vol 27 no 9 pages 57-59 See chemical abstract	1-3, 6, 13
X	GB 1207326 (UNILEVER LIMITED) 30 September 1970 See page 2 lines 1-3, 15-21, 35-42, 50-61, 67-117, 125-128; page 3 lines 1-19	1-46
X	STN FILE CA Abstract No.12:13041 & DE ASTIS, G, Ann. Chim. Applicata (1918) vol 9 pages 155-241 See chemical abstract: item (6) of "Citric Acid..." ie lines 180-181 of the abstract	1-6, 13
P,X	BERGERON, C et al, "Stabilization of caffeic acid derivatives in Echinacea purpurea L. glycerin extract", Journal of Agricultural and Food Chemistry, (3 July 2002) vol 50 no 14 pages 3967-3970 See abstract, page 3967 column 1 paragraphs 2-3, column 2 paragraph 1, page 3969 column 2 paragraphs 2-4, Figures 3-6, page 3970 column 1 paragraphs 2-3	1-5, 10-46

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

PCT/NZ02/00230

This Annex lists the known "A" publication level patent family members relating to the patent documents cited in the above-mentioned international search report. The Australian Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

Patent Document Cited in Search Report			Patent Family Member				
US	6004559	US	5837252	US	5945106	US	6039955
		US	6240964	AU	48956/97	WO	9815184
		AU	97754/98	WO	9917609	AU	37358/97
		BR	9710123	CA	2258761	EP	907856
		KR	2000022524	US	6125890	WO	9800664
US	5427806	AU	19822/95	WO	9604802		
GB	1207326	BE	721048	CH	506953	DE	1792553
		ES	358250	FR	1580731	NL	6813247
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